IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No. : 10/660,382 Confirmation No.: 6022
Applicant : Graetz et al.
Filed : September 10, 2003
TC/A.U. : 1795
Examiner : Lee, Cynthia K.
For : High-Capacity Nanostructured Silicon and Lithium Alloys Thereof
Docket No. : 26-06
Customer No.: 23713

MAIL STOP AMENDMENT
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION OF RACHID YAZAMI UNDER 37 CFR 1.132

Sir:

Rachid Yazami hereby declares as follows:

1. I, Rachid Yazami, am an inventor of the above-identified U.S. Patent Application No. 10/660,382, filed on September 10, 2003.

2. I am presently a Research Director at Centre National De La Recherche Scientifique (C.N.R.S.) and a Visiting Associate in Chemistry at the California Institute of Technology.

3. I have experience and expertise in the field of electrochemistry and material science, including silicon electrodes and lithium electrochemical cells.

4. I have reviewed the Office Action of September 17, 2009 for U.S. Patent Application No. 10/660,382 and the references cited therein, including Abstract 257 of The 11th International Meeting on Lithium Batteries in Monterey, CA on June 23-28, 2002, entitled 'Li Insertion/Extraction Reaction

of a Si Film Evaporated on a Ni Foil' (Takamura et al.); and U.S. Patent Publication 2002/0048705 (Park et al.).

5. The Takamura et al. and Park et al. references **do not** disclose or otherwise suggest a silicon electrode having a silicon oxide outer layer that is 18.5% to 50% SiO_2 by weight. In addition, this property cannot be reasonably inferred as inherent to the electrodes and electrochemical cells disclosed in these references given the technical information provided in Takamura et al. and Park et al.

6. Incorporation of a silicon oxide outer layer having a weight percentage of 18.5% to 50% SiO_2 in the nanostructured silicon electrodes of the present invention provides important attributes fundamental to the electrochemical properties and the performance of the claimed electrodes in a secondary electrochemical cell. These attributes include providing an effective solid electrolyte interphase (SEI) layer and overall charge and discharge capacity critical to use of the nanostructured material as an electrode for a secondary electrode chemical cell.

7. The lower limit of **18.5% wt%** of the SiO_2 layer and its position as an **outer layer** is critical as it provides a nanostructured material (e.g., nanofilm or nanoparticles) having a SiO_2 layer with a thickness large enough to provide properties critical for implementation of the nanostructured silicon material as an electrode for a secondary electrochemical cell. For example, a SiO_2 outer layer having a percentage by weight of 18.5% or greater prevents the battery from significantly self-discharging during use in an electrochemical cell. The upper limit of **50% wt%** of the SiO_2 outer layer is also critical as it provides a nanostructure material with a charge and discharge capacity high enough for it to function as an electrode for a battery application, such as in a secondary

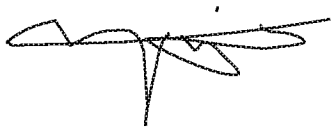
electrochemical cell. For example, the actual capacity of the nanostructured silicon electrode is expected to decrease linearly with weight percentage of SiO_2 as provided by the expression:

$$Q(mAh/g)= 4212-22.46(wt\ \%\ SiO_2);$$

wherein a 50% wt% of SiO_2 is expected to provide a capacity (Q) equal to 3089mAh/g, which is a capacity high enough for battery applications, such as use in a secondary electrochemical cell. Accordingly, a specific weight percentage of 18.5% to 50% wt% of SiO_2 in the nanostructured silicon electrodes is critical for providing an electrode for a secondary electrochemical cell with low self-discharge rate and high storage capacity. Justification of the expression above is given in Exhibit 1, attached to the present declaration.

8. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the above-identified U.S. Patent Application No. 10/660,382 or any patent issuing thereon.

Date: March 17, 2010



Rachid Yazami

Exhibit 1

Justification of the Expression:
$Q(mAh/g)= 4212-22.46(wt\ \%\ SiO_2)$

Assuming the silicon based electrode material has a formula: Si_pO.

In the formula Si_pO silicon is present as pure element Si in the bulk and as SiO_2 outer layer.

Accordingly, Si_pO can be expressed by: $Si_{p-1/2}(SiO_2)_{1/2}$

The wt % of SiO_2 'σ' in the $Si_{p-1/2}(SiO_2)_{1/2}$ formula is:

$$\sigma = wt.\%SiO_2 = 100\frac{30}{28p+16} \quad (1)$$

(O=16, Si=28, SiO_2=60)

Therefore,

$$p = \frac{3000-16\sigma}{28\sigma} \quad (2)$$

When used as an anode material Si_pO reacts with lithium to form $Li_{4.4}$ Si and Li_2O according to:

$$Si_pO + (4.4p+2)Li \rightarrow pLi_{4.4}\,Si + Li_2O \quad (3)$$

Lithium involved in $Li_{4.4}$ Si is the only one useful for the reversible capacity of the electrode:

$$pLi_{4.4}\,Si \leftrightarrow 4.4pLi + pSi \quad (4)$$

Therefore, the reversible specific capacity of the Si_pO electrode is:

$$Q\left(\frac{mAh}{g}\right) = \frac{4.4pF}{3.6(28p+16)} \quad (5)$$

F=Faraday number=96500C

Combining Eqns. (2) and (5) gives the Expression:

$Q(mAh/g)= 4212-22.46\ \sigma$ (σ= wt % of SiO_2)